Degenerate multiwave mixing and phase conjugation in silicon

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We present a detailed theory and experiment on degenerate-multiwave-mixing-mediated beam amplification in the semiconductor silicon. The theory accounts for several important factors, such as coupling to higher-order diffractions, intensity-dependent self- and mutual-phase modulations, losses, phase matchings, intensity ratio among the input beams, etc. In the silicon case under study, involving the generation of electron-hole pairs by 1.06-μm laser pulses, the intensity-dependent absorption loss and the temporal and/or spatial intensity profile of the input laser beam are also explicitly accounted. The experimental results for probe-gain dependences on several parameters are in good agreement with the theory. The important role played by the side diffractions in mediating the amplification of the probe is also apparent in our experiment on phase conjugation of the probe beam, where a strong dependence of the phase-conjugation reflection on the pump- to probe-beam ratio is observed, similar to the probe-gain dependence.

I. INTRODUCTION

Degenerate optical wave mixings in highly nonlinear media, such as photorefractive crystals, semiconductors, liquid crystals, and organic materials have been vigorously studied in the past few years.[1–7] Among the various processes that are of fundamental and applied importance, phase-conjugation reflection and amplification of a probe beam have been well studied. For semiconductor materials, a comprehensive review of the fundamental mechanisms and experiments may be seen in the review article by Jain and Klein.[8] More recently,[9–11] degenerate four-wave mixings in semiconductors have been studied in superlattices, multiple-quantum-well structures, doped glasses, etc. for probing the extraordinarily large optical nonlinearities associated with quantum confinement and materials and structural dependences. In this paper, these optical-mixing processes in semiconductors are revisited in the light of recently observed multiwave mixing *(involving the incident pump and probe beams, and one or more diffracted beams)* effects in silicon.[12,13] Studies in silicon[12–14] and other nonlinear materials such as liquid crystals[15] have demonstrated that in optically thin media, the side diffractions, which are often regarded as weak beams, can in fact lead to substantial contribution to the amplification of the probe beam. *These multiwave-mixing effects are expected to also affect the phase-conjugation reflectivity.* In view of the rapid emergence of a large number of semiconductor thin films with extraordinarily large optical nonlinearity, where these multiwave-mixing effects are expected to play even more significant roles in probe-amplification and phase-conjugation processes, we present here a detailed theory along with experimental results on the factors involved. Specifically, we point out the importance of accounting for intensity-dependent self- and mutual-phase modulations, couplings with higher-order diffraction beams, pump- to probe-beam ratio, medium losses, etc. These factors are important in obtaining both a correct fit of the experimental results and extracting material parameters (e.g., third-order nonlinear susceptibility) using phase conjugation and in explaining the observed wave-mixing effects. These factors have hitherto been neglected in most theories[13,16] on this subject of wave-mixing-induced probe amplification.

In Sec. II, we will present a general theory of optical wave mixing involving a strong pump beam and a probe beam and two diffracted beams. This theory is then extended in Sec. III to the case involving another pump beam counterpropagating to the first pump beam.

In Sec. IV, the basic mechanisms of the absorption of 1.06-μm laser pulses by silicon and the resulting intensity-dependent refractive-index change and losses caused by nanosecond Gaussian laser pulses are discussed. Also, the explicit dependence of the photogenerated carriers and attenuation by nanosecond laser pulses with a spatially Gaussian profile are derived. Some of the new results are discussed in Sec. V, where experimental results with silicon and neodymium-doped yttrium aluminum garnet (Nd:YAG) lasers (1.06 μm) are also presented. In the Conclusion, we summarize the general results of and insights gained from this study.

II. FORWARD-MULTIWAVE-MIXING EFFECT

Figure 1(a) depicts schematically the interaction of a pump and a probe beam (coherent with respect to each other) interacting in a nonlinear medium. Owing to the optical intensity-dependent refractive index of the nonlinear medium, a refractive-index grating is generated via the interference of the two incident beams. In general, this interaction produces several side diffractions (beams 3, 4, etc.) depending on the magnitude of the index modulations generated by the pump and the probe, as well as the interaction length within the nonlinear medium. The magnitude of these diffractions, in comparison to the input beams, are usually small, and they are often neglected in the usual theory dealing with the amplification, of the

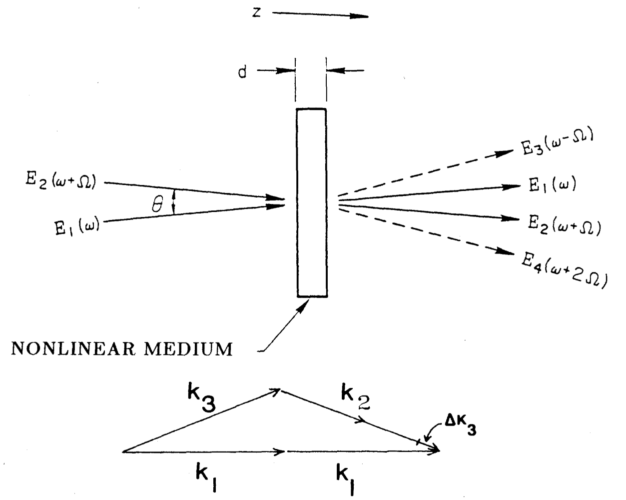

FIG. 1. (a) Schematic diagram of the multiwave-mixing geometry involving a strong pump beam, 1, and a weak probe beam, 2. Beams 3, 4, etc. are diffractions from the pump-probe-induced refractive-index grating. (b) Wave-vector diagram for the scattering of the pump beam from the pump-diffracted-beam grating into the probe-beam direction.

probe by the pump. However, in Kerr-like media, our recent theory[17] and experiments[15] have shown that one has to include the effect of the diffracted beam coupling to the pump beam, in order to explain the amplification of the probe. This process is depicted in Fig. 1(b), which shows the wave-vector diagram corresponding to the scattering of the pump beam (1) from the pump-diffracted beam-generated grating, into the direction of the probe beam. Because there is a corresponding process involving the other diffracted beam (4) which scatters the probe beam into the pump-beam direction, the gain in intensity of the probe will occur if it is weaker than the pump.

From this point of view, one can say that the diffracted beam 3, in conjunction with the pump, is a source for gain or amplification for the probe beam, whereas the diffracted beam 4 is a source of loss for the probe beam. This picture is qualitatively correct, but it misses out on the crucial role played by various optical phase factors involved in the process. In Ref. 17, where we presented a complete theory involving a complex optical field (E), the effects played by the phases are not apparent. In this paper, these equations are reexpressed in terms of the intensity (I) and the phase (ϕ), and their respective roles becomes more apparent.

Following the notations of Ref. 17, and with the definition for the complex field amplitude E_j,

$$\left(\tfrac{1}{2}\sqrt{\epsilon/\mu}\right)^{1/2}E_j=\sqrt{I_j}\exp(i\phi_j),\quad j=1,2,3,4 \tag{1}$$

etc., for all the optical fields involved, the relevant equations may be written as follows:

$$\begin{aligned}\frac{dE_1}{dz}=&-ig'(|E_1|^2+2|E_2|^2+2|E_4|^2+2|E_3|^2)E_1\\&-ig'\{E_2^2E_4^*\exp[i(-\Delta k_3 z)]+E_2^*E_3E_4\exp(2i\,\Delta k_3 z)+E_1^*E_2E_3\exp(i\,\Delta k_3 z)\}-\tfrac{1}{2}\alpha E_1\ ,\end{aligned} \tag{2}$$

$$\begin{aligned}\frac{dE_2}{dz}=&-ig'(|E_2|^2+2|E_1|^2+2|E_3|^2+2|E_4|^2)E_2\\&-ig'[E_1^2E_3^*\exp(-i\,\Delta k_3 z)+E_1E_4E_2^*\exp(i\,\Delta k_3 z)+E_3E_4E_1^*\exp(2\,\Delta k_3 z)]-\tfrac{1}{2}\alpha E_2\ ,\end{aligned} \tag{3}$$

$$\begin{aligned}\frac{dE_3}{dz}=&-ig'(|E_3|^2+2|E_1|^2+2|E_2|^2+2|E_4|^2)E_3\\&-ig'\{E_1^2E_2^*\exp[-i(+\Delta k_3 z)]+E_1E_2E_4^*\exp(-2i\,\Delta k_3 z)\}-\tfrac{1}{2}\alpha E_3\ ,\end{aligned} \tag{4}$$

$$\begin{aligned}\frac{dE_4}{dz}=&-ig'(|E_4|^2+2|E_1|^2+2|E_2|^2+2|E_3|)E_4\\&-ig'\{E_2^2E_1^*\exp[i(-\Delta k_3 z)]+E_1E_2E_3^*\exp(-2i\,\Delta k_3 z)\}-\tfrac{1}{2}\alpha E_4\ ,\end{aligned} \tag{5}$$

where α is the intensity absorption constant (cf. Sec. IV) and Δk_3 is given by

$$\Delta k_3=|2\mathbf{k}_1-\mathbf{k}_2-\mathbf{k}_3|\ , \tag{6}$$

and

$$\phi_{ij}(z)=\phi_i(z)-\phi_j(z)\ , \tag{7}$$

and the coupling parameter g' is given by

$$g'=\frac{\omega^2 n_0 n_2}{2k_z c^2}\ , \tag{8}$$

assuming that the medium has an intensity-dependent refractive index n of the form

$$\begin{aligned}n&=n_0+n_2(E)|E|^2\\&=n_0+n_2(I)I\ ,\end{aligned} \tag{9}$$

where n_0 is the refractive index in the absence of the optical fields. The nonlinear coefficient $n_2(I)$ is related to $n_2(E)$ by $n_2(E)=(\frac{1}{2}\sqrt{\bar{\epsilon}/\mu})n_2(I)$. This form of intensity dependence for the refractive index is of course an approximation; in actual experiments (cf. experimental parameters quoted in Sec. V), the laser pulse used is short or comparable to the lifetime of the grating (dominated by carrier diffusion lifetime). A more accurate way of writing n will be to express it in terms of a time integral of $E(t)E(t)$ (cf. Ref. 8). To incorporate this nonlocal (in time) dependence in the coupled wave equations will involve a tremendous computation time and masks the point we are trying to make here, namely, to point out the *dependences* of the probe-beam amplification on several factors that appear important. As is often done (cf., e.g., Ref. 8), for practical cases where one is interested in or is experimentally measuring the response at the temporal peak of the laser pulse, the expression for n given in (9) may be used for estimating n_2. In the above equations, k_z is the wave-vector component in the z direction, and $\bar{\epsilon}$ and μ the dielectric constant and permeability of the material, respectively. With the following redefinitions:

$$\Phi_1(z)=\phi_{12}(z)+\phi_{13}(z)-\Delta k_3 z\ , \tag{10}$$

$$\Phi_2(z)=\phi_{12}(z)+\phi_{42}(z)+\Delta k_3 z\ , \tag{11}$$

$$\Phi_3(z)=\phi_{31}(z)+\phi_{42}(z)+2\,\Delta k_3 z\ . \tag{12}$$

Equations (2)–(5) become

$$\begin{aligned}\frac{dI_1}{dz}&=-\alpha I_1-4gI_1\sqrt{I_2I_3}\sin\Phi_1-2gI_2\sqrt{I_1I_4}\sin\Phi_2\\&\quad+4g\sqrt{I_1I_2I_3I_4}\sin\Phi_3\ ,\\\frac{dI_2}{dz}&=-\alpha I_2+2gI_1\sqrt{I_2I_3}\sin\Phi_1+4gI_2\sqrt{I_1I_4}\sin\Phi_2\\&\quad+4g\sqrt{I_1I_2I_3I_4}\sin\Phi_3\ ,\end{aligned} \tag{13}$$

$$\frac{dI_3}{dz}=-\alpha I_3+2gI_1\sqrt{I_2I_3}\sin\Phi_1-4g\sqrt{I_1I_2I_3I_4}\sin\Phi_3\ , \tag{14}$$

$$\frac{dI_4}{dz}=-\alpha I_4-2gI_2\sqrt{I_1I_4}\sin\Phi_2-4g\sqrt{I_1I_2I_3I_4}\sin\Phi_3\ , \tag{15}$$

and the equations for the phases (11)–(15) become

$$\begin{aligned}\frac{d\Phi_1}{dz}&=-g(I_1+2I_2+2I_3+2I_4)-gI_2\sqrt{I_4I_1}\cos\Phi_2\\&\quad-2g\sqrt{I_2I_3}\cos\Phi_1-2g\sqrt{I_2I_3I_4/I_1}\cos\Phi_3\ ,\end{aligned} \tag{16}$$

$$\begin{aligned}\frac{d\Phi_2}{dz}&=-g(I_2+2I_1+2I_3+2I_4)-gI_1\sqrt{I_3/I_2}\cos\Phi_1\\&\quad-2g\sqrt{I_1I_4}\cos\Phi_2-2g\sqrt{I_1I_3I_4/I_2}\cos\Phi_3\ ,\end{aligned} \tag{17}$$

$$\begin{aligned}\frac{d\Phi_3}{dz}&=-g(I_3+2I_1+2I_2+2I_4)-gI_2\frac{\sqrt{I_2}}{I_3}\cos\Phi_1\\&\quad-2g\frac{\sqrt{I_1I_2I_4}}{I_3}\cos\Phi_3\ ,\end{aligned} \tag{18}$$

$$\begin{aligned}\frac{d\phi_4}{dz}&=-g(I_4+2I_1+2I_2+2I_3)-gI_2\frac{\sqrt{I_1}}{I_4}\cos\Phi_2\\&\quad-2g\frac{\sqrt{I_1I_2I_3}}{I_4}\cos\Phi_3\ ,\end{aligned} \tag{19}$$

where the coupling constant g is given by $g=(g'/2)\sqrt{\bar{\epsilon}/\mu}$. Notice that the coupling and energy interchanges among the four beams are intricately governed both by the intensities (I_1,I_2,I_3,I_4) as well as the phase factors (Φ_1,Φ_2,Φ_3). Our extensive numerical solution[17] of the above equations have shown that, *ignoring any of these phase factors, or the phase mismatches (Δk_3), or other factors such as loss, and self- and mutual-phase-modulation terms* [c.f. terms grouped on the first line on the right-hand side of Eqs. (17)–(19)] *will lead to erroneous results on the exiting beam intensities.* In particular, without including the fourth beam [i.e., using only a three-wave- (pump, probe, and diffracted-beam 3) mixing model], one cannot explain the dependence of the probe-beam amplification on the pump- to probe-beam intensity ratio. The three-wave model is correct only in the limit of very large pump- to probe-beam ratio, which inevitably breaks down once the probe beam experiences substantial gain. For example, the incident pump- to probe-beam ratio may be 100 to 1. The probe could experience a gain of 20 (which has been observed) and thus effectively the pump-to-probe ratio is reduced to 5 to 1 at the exit plane. Under these conditions, the inclusion of the fourth wave is absolutely necessary in accurately accounting for the lowering of the probe gain due to its (the probe's) diffraction into the fourth-beam direction.

III. EFFECT ON PHASE CONJUGATION

Since the probe beam can experience very large gain, the phase-conjugated reflection of the probe beam will also be considerably changed by coupling to the higher-order diffractions. Figure 2 shows the relevant beams involved in this process. Because this situation involves input beams at two planes ($z=0$ and $z=d$, the sample thickness) and unknown values at these planes for the diffracted and conjugated beams, numerical solution of this problem is very complicated, and we reserve it for a separate article elsewhere[18] dealing with the theory of optical phase conjugation in a general nonlinear media. It suffices to point out here that because the phase-conjugation reflection depends on the probe-beam intensity, which in turn depends on the pump- to probe-beam ratio, one signature of the effect of these side diffractions is the dependence of the phase-conjugation reflectivity on the probe intensity (i.e., the phase-conjugation reflectivity should vary in a manner almost identical with the probe-beam-gain dependence on the pump- to probe-beam ratio). This is indeed shown to be the case in our *experimental results* reported in a later section of this paper.

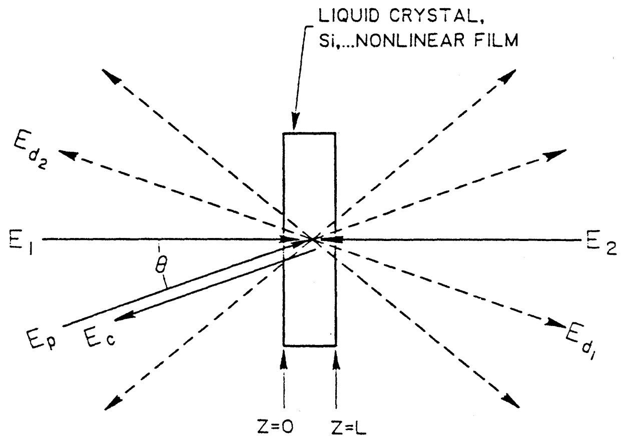

FIG. 2. Schematic diagram of the phase-conjugation process involving the strong pump beam 1, the weak probe beam P, and a strong reconstruction beam, 2, counterpropagating to beam 1. The generated phase-conjugated signal propagates backward along the probe beam. Diffractions are generated in the forward, as well as the backward, directions.

IV. SILICON NONLINEARITY AND INTENSITY-DEPENDENT LOSSES INDUCED BY 1.06-μm LASER PULSES

The preceding discussions are centered on what we may term as the optical part of the problem. To accurately describe the experimental results in silicon (or for that matter, any nonlinear media), the peculiarities of the nonlinear mechanism must be accounted for. As discussed in a previous short communication, the primary nonlinear-optical response in silicon is associated with the generation of free carriers by indirect valence to conduction-band transition. The carrier concentration $N(t)$ is related to the optical intensity $I(t)$ by

$$\frac{\partial N(t)}{\partial t}=\frac{\alpha(T)I(t)}{\hbar\omega}\ , \tag{20}$$

where $\alpha(T)$ is the temperature-dependent linear absorption constant. Since the laser pulse is short, the recombination and diffusion terms are neglected in Eq. (20). From the Drude model,[8] the change in refractive index is given by

$$\Delta n=\frac{-e^2\tau\alpha(T)\sqrt{\pi}}{4nm_{e\text{-}h}\omega^2\epsilon_0\hbar\omega}I(t)\ , \tag{21}$$

where τ is the laser pulse length and $m_{e\text{-}h}$ is the electron-hole mass, and $\hbar$ is Planck's constant divided by 2π; as a result of the photoabsorption, the temperature of the silicon rises. The temperature T is related to the free-carrier absorption constant $\sigma_{e\text{-}h}$ by

$$C\frac{\partial T}{\partial t}=\sigma_{e\text{-}h}(T)N(t)I(t)\ , \tag{22}$$

where C is the heat capacity per unit volume.

Because of the interrelationship between $I(t)$, $N(t)$, $T(t)$, $\alpha(T)$, and $\sigma_{e\text{-}h}(T)$ through Eqs. (20)–(22), and the relationship between the various beams' intensity (pump, probe, and diffracted) through Eqs. (13)–(19), and various transmitted beam parameters of interest (e.g., probe-beam intensity) must be solved from Eqs. (13)–(19) and (20)–(22) in a self-consistent manner. Our numerical solution to this problem is solved by accounting for the temperature rise of the silicon due to the strong pump beam alone. This is correct for the experiments involving the mixing of a strong pump beam and a weak probe. (Typically, the pump- to probe-beam intensity ratio is larger than 10.) Even under this approximation, the numerical solution is complicated by the fact that the pump laser pulse is Gaussian in time and space. The laser pulse is of the form $I(t)=I_0\exp[-(t/\tau)^2]$, where I_0 is the peak-beam-center intensity and τ the pulse duration.

For this Gaussian laser pulse, the rise in temperature ΔT is given by

$$\Delta T(t)=\frac{\alpha(T)\sigma_{e\text{-}h}(T)I_0^2\tau^2\pi}{8C\hbar\omega}\times[\mathrm{erf}^2(t/\tau)+\mathrm{erf}(t+\tau)+1]\ , \tag{23}$$

and the temperature-dependent linear absorption constant $\alpha(T)$ is given by

$$\alpha(T)=\left[\frac{T}{172.3}\right]^{4.25}\ \mathrm{cm}^{-1} \tag{24}$$

(cf. Ref. 19) and the free-carrier absorption constant by

$$\sigma_{e\text{-}h}=1.7\times10^{-20}T\ \mathrm{cm}^2 \tag{25}$$

(cf. Ref. 19). Equations (23)–(25) are used in the numerical computation.

Numerically, we perform a three-dimensional space and time self-consistent solution of Eqs. (20)–(22) for a pump beam of input intensity up to 10^7 W/cm^2. These values for the pump-beam intensities for various beam penetrations into the nonlinear beam are then compiled, and used in the solution[20] of Eqs. (13)–(19). These transmitted power dependences on the input intensity have been experimentally verified in a separate study.[20] In the present context, these numerically calculated values for the pump beam provide a more realistic simulation of the loss suffered by all of the laser beams in tranversing the silicon sample. In our experiment, the *peak intensity* of the transmitted probe is detected. Therefore, in our numerical calculation, we use the numerical data compiled from these calculations for the peak intensities.

For the purpose of easy reference, we list the numerical values of various physical parameters of the silicon wafer used, which range in thickness from 100 to 500 μm. The refractive index n of Si is 3.56, the diffusion constant $D\sim19$ cm^2/s;[21] the effective electron-hole mass $m_{e\text{-}h}=0.16m_e$, the heat capacity $C=0.7\ \mathrm{J\,g^{-1}k^{-1}}$, the initial temperature $T=300$ K, the laser wavelength $\lambda=1.06\ \mu$m, and the pulse duration $\tau=20$ ns. Crossing angles between the pump and probe beams are varied from 15×10^{-3} to 45×10^{-3} rad; the laser is p polarized and incident on the silicon wafer at the Brewster angle to reduce reflection loss. For the crossing angles (between the pump and probe) used, the grating constant Λ ranges from 70 to 23 μm. This corresponds to grating diffusion time constants $\tau_g = \Lambda^2/D$ of about 111–6.8 ns, respectively.

V. EXPERIMENTAL RESULTS AND DISCUSSIONS

We have performed both a forward-wave-mixing probe-amplification study and optical phase-conjugation studies.

The laser used is a single longitudinal mode Nd:YAG 1.06-μm laser that is Gaussian in time and transverse spatial profile. The experimental parameters are generally the same as that described in the last paragraph of the preceding section. The experimental details for the forward probe-beam-amplification effect have been reported in a previous short communication. Some new insights will be discussed. In general, we found that the observed probe gain as a function of the input pump intensity can be explained quantitatively only by the kind of self-consistent solution of the wave-mixing equations (13)–(19) and the coupled temperature-free carrier-generation equations (20) and (21) discussed above. In Ref. 12, for example, we showed that the observed probe gain (circles) can be nicely fitted by our theory (solid line *a*) from which we can extract a value of $n_2(I) = 3.5 \times 10^{-12}$ SI units (m^2/W) or, in cgs units, a third-order nonlinear susceptibility $\chi^{(3)}$ of 7.7×10^{-7} esu.

The monitoring of the principal diffraction beam 3 also provides further insight into this wave-mixing result. Figure 3 shows a plot of the observed diffracted-beam-3 energy versus the pump energy at a fixed pump-probe ratio of 61 and a crossing angle in air of 16×10^{-3} rad. In our experiment, this ratio is chosen so that even at the highest pump power used, the *higher-order diffracted beam, 4, is still negligible small.* At low pump energy, we observed that the diffracted-beam power obeys a cubic power law, in agreement with a simple four-wave-mixing process. The observed diffracted-beam power, however, began to fall considerably below this cubic dependence at high pump power, again due to the severe loss caused by the free-carrier-attenuation effect on the laser. The diffracted-beam power actually "saturates" and begins to drop at an input pump energy fluence of 200 mJ/cm^2 (equivalently, an intensity of 6 MW/cm^2), in close agreement with the single (pump) beam transmission (whole beam-power) curve given in Fig. 3.

As we mentioned earlier, the amplification of the probe beam via the multiwave-mixing effect is highly dependent on the pump- to probe-beam ratio. This was conclusively shown in Ref. 12. In general, at pump- to probe-beam ratios close to unity, the presence of a "strong" probe beam creates significantly more free carriers, and therefore increases the loss experienced by both beams. This increased loss is responsible for the lower values for the experimentally observed gain than the theoretical prediction. Put another way, our theory that accounts for the loss caused by free-carrier generation by the pump beam alone is not adequate. At higher pump- to probe-beam ratio, i.e., the probe beam is much weaker, the theory provides a better description of the experimental results.

In all experimental studies, the probe gain saturates to a fixed value at high pump- to probe-beam ratio. This effect can in fact be proven analytically: if the pump- to probe-beam ratio is extremely large, i.e., we have a very weak probe beam, then only the diffraction beam 3 will be significant. The four-beam-mixing effect then becomes a simple three-beam-mixing effect [cf. Fig. 1(b)], and the gain of the probe beam is simply a function of the pump-beam intensity alone.[22] From this point of view, the *dependence of the probe gain on the pump- to probe-beam ratio* is a manifestation of the effect due to the diffracted beam 4.

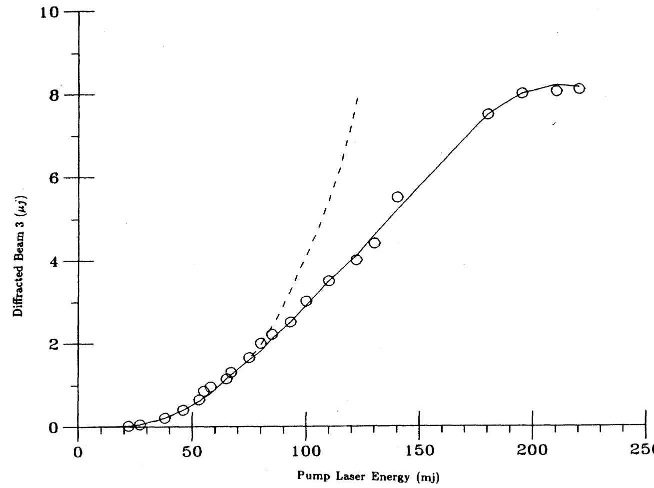

FIG. 3. Observed diffracted-beam power as a function of the pump-beam intensity for a fixed pump- to probe-beam ratio. Dotted line shows the three-beam wave-mixing theory without accounting for the intensity-dependent loss.

The amplification of the probe gain in this multiwave-mixing effect is crucially dependent on the presence of the diffracted beams. On the other hand, the intensity of the diffracted beam depends critically on the phase mismatch $\Delta k_3 l$, where l is the interaction length inside the nonlinear medium. For a given crossing angle θ between the pump and the probe beam, the phase mismatch $\Delta k_3 l \approx k\theta^2 l$ can become appreciable ($\approx \pi$) for $\theta > (\pi/kl)^{1/2} = \theta_\pi$. For the 0.5-mm-thick sample used, the interaction length inside the crystal is given by $l = (0.5 \text{ mm})/\cos\phi$, where ϕ is the angle made by the laser-propagation direction with the normal to the plane of the sample. The angle ϕ is related to the angle in air, ϕ_{air}, by $\sin\phi_{\text{air}} = 3.56 \sin\phi$. In our experiment, ϕ_{air} is chosen to be the Brewster angle to minimize reflection loss (i.e., $\tan\phi_{\text{air}} = 3.56$). This gives $\phi_{\text{air}} = 74.3°$, $l = 0.52$ mm, and $\theta_\pi(\text{in air}) \approx 3°$, i.e., we expect the diffracted beam to be quenched by the phase mismatch for a pump-probe crossing angle θ greater than 3°. Experimentally, this shows up as a sharp diminishing in the probe gain as a function of the crossing angle, as plotted in Fig. 4. At $\theta > 0.04$ rad (2.4°) there is essentially no measurable probe gain. On the other hand, a gain of 4 is observed for $\theta \approx 0.02$ rad. For these two values of crossing angle, the grating diffusion time constants are on the order of 25 ns (for $\theta = 0.02$ rad) and 6 ns (for $\theta = 0.04$ rad). Hence, if the laser-pulse length is on the order of 20 ns, one would

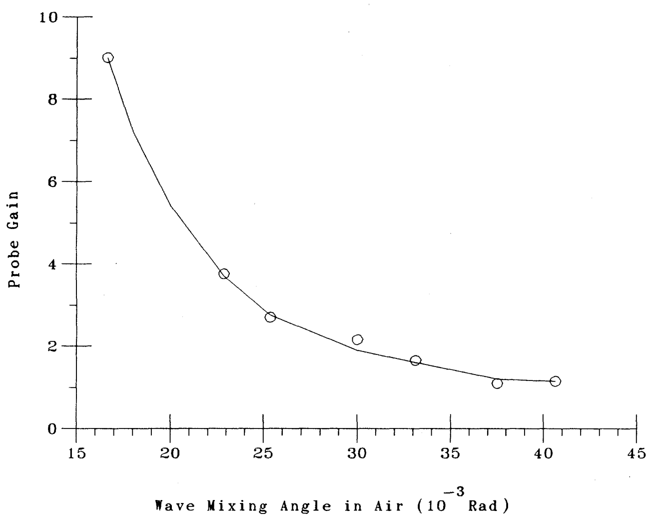

FIG. 4. Experimentally observed probe-beam gain as a function of the pump-probe crossing angle in air. The pump-beam energy is 20 mJ and the probe beam energy is 7.7 μJ (pump- to probe-beam ratio of 2600).

also expect the grating diffusion to contribute somewhat to the drop in the gain.

As we remarked earlier, this multiwave mixing, which affects the magnitude of the probe-beam intensity in the nonlinear medium, clearly will affect the magnitude of the phase-conjugation reflection of the probe beam. We have performed a typical optical-wave-front-conjugation experiment with a probe beam of variable intensity ratio with respect to the pump beams. Figure 5 shows the experimental setup. The 1.06-μm laser nano second pulsed from the Nd:YAG laser are divided into two equal pump beams that are counterpropagating to each other. A weak probe overlaps with the forward-propagating pump beam on the sample. The phase-conjugated reflection of the probe beam is monitored as a function of the probe beam as it is varied by a variable-beam attenuator (VBA), while the pump beams are kept constant. The silicon wafer used is the same as that reported in the previous experiment. The two pump-beam energies are 4 mJ, and the probe-beam energy is varied such that the pump- to probe-beam ratio varies from 7 to 500.

Because of the presence of an additional strong pump beam, the intensity-dependent attenuation effect (due to the laser-induced electron-hole plasma) on the beam in

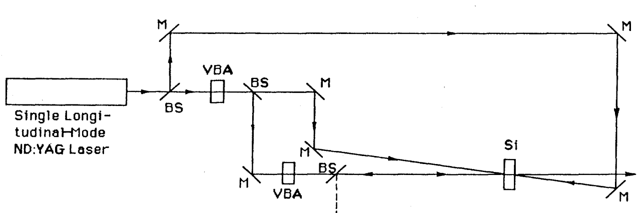

FIG. 5. Schematic diagram of the experimental setup for studying the dependence of the phase-conjugation reflection of the probe beam on the pump- to probe-beam ratio.

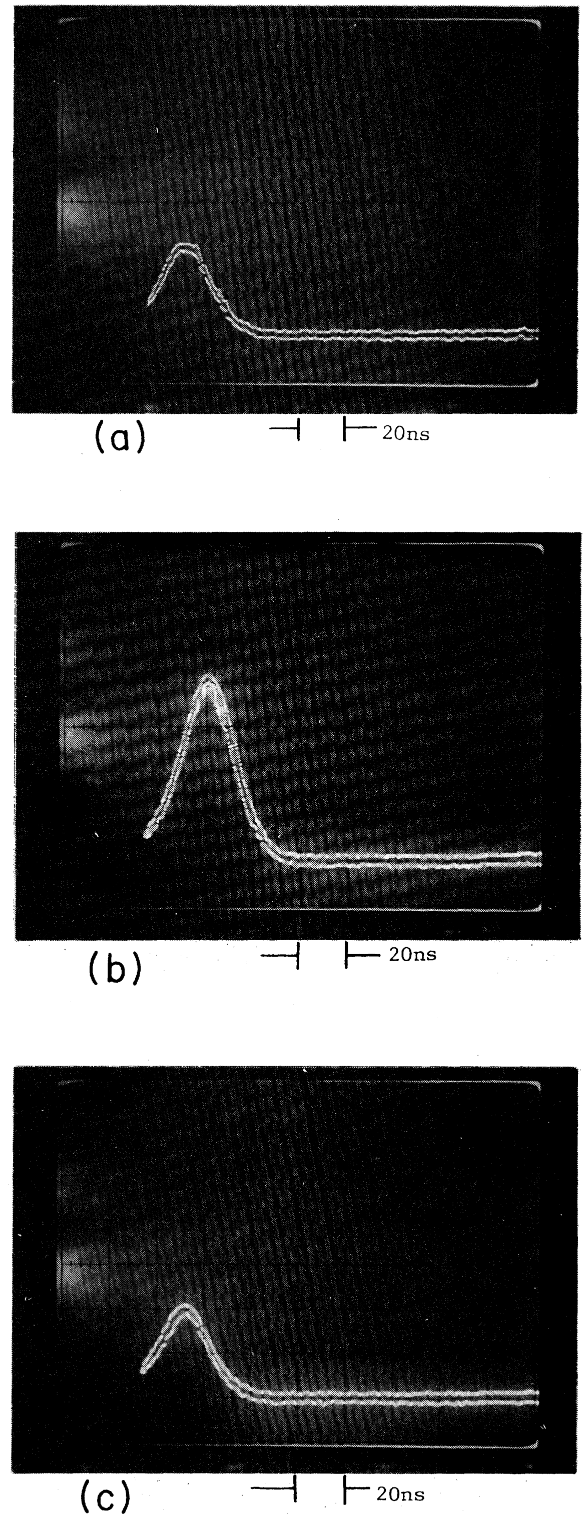

FIG. 6. (a) Oscilloscope trace of the probe pulse alone. The double trace is an artifact of the transient used in the pulse detection. (b) Oscilloscope trace of the probe beam in the presence of the forward-propagating pump beam (I_f) alone, showing an amplification of the probe beam. (c) Oscilloscope trace of the probe beam with both pump beams on (I_f and I_b), showing diminished gain.

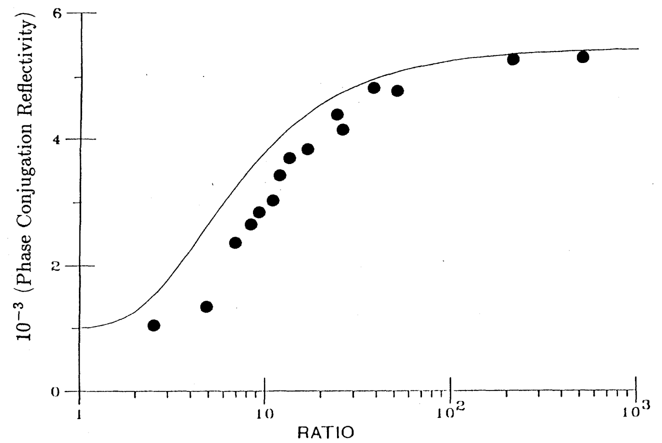

FIG. 7. Experimentally observed phase-conjugation reflectivity of the probe beam as a function of the pump- to probe-beam ratio. The solid line is a plot of the corresponding probe-gain dependence on the beam ratio.

traversing the nonlinear medium becomes very pronounced. This may be seen by monitoring the transmitted probe beam. Figure 6(a) is an oscilloscope trace of the probe pulse with both pump beams blocked. Figure 6(b) shows the probe pulse with the forward pump beam on, clearly exhibiting an amplification effect. However, when the counterpropagating beam is also present, the probe-beam amplification is considerably reduced owing to the extra free-carrier generation by the counterpropagating pump beam, as shown in Fig. 6(c).

The presence of both pump beams is, of course, needed for the generation of the phase-conjugated reflection of the probe beam. In spite of the large losses suffered by the beams, the multiwave-mixing effect (which is responsible for the amplification of the probe beam) is also evident in the phase-conjugated probe reflection. Figure 7 (solid dots) shows the experimentally observed phase-conjugation reflectivity (intensity of probe reflection divided by intensity of incident probe) as a function of the pump- to probe-beam intensity ratio. The reflectivity varies from about 1.6×10^{-3} at small beam ratio to about 5 times this value at large beam ratio. When the reflectivity is plotted as a function of the beam ratio, as shown in Fig. 7, it is strikingly similar to the probe-beam–gain dependence on the beam ratio. (The solid line in Fig. 7 is a theoretical plot *of the probe gain* as a function of the beam ratio and is adjusted so that the high-ratio portions fits the experimental result for comparison purposes.) The similarity in the dependence on the ratio clearly shows that the underlying forward-multiwave-mixing effect responsible for the amplified probe is also responsible for the increased phase-conjugation reflection, as the pump- to probe-beam ratio is increased.[23] At a large value of the ratio, i.e., very low probe-beam intensity, again the phase-conjugation reflectivity approaches a "saturated" value, which corresponds to a simple degenerate four-wave-mixing process similar to the probe-gain effect discussed earlier.

VI. CONCLUSION

We have presented a theory of multiwave-mixing-mediated 1.06-μm laser-probe-beam amplification in silicon. The coupled equations are solved numerically. The theory accounts for the various roles played by beam intensity, beam ratio, phase modulation, intensity-dependent losses, and relevant physical mechanisms such as temperature, free-carrier generation, etc. in silicon. Experimentally observed probe-gain diffracted-beam and various other dependences are in good agreement with theoretical expectation. This theory, which combines the wave-mixing theory of Khoo and Liu and the theory for spatially and temporally Gaussian 1.06-μm laser-pulse interaction with silicon, provides us with new insights into degenerate wave mixing in silicon, particularly on the role of the diffracted beam on the probe-beam gain, and its subsequent effect on the phase-conjugated probe reflection. We have shown that because of the effect of the diffracted beam, the phase-conjugated probe reflection is highly dependent on the relative strength of the probe beam with respect to the pump. These observations are important in any material nonlinear-response characterization using degenerate-wave-mixing process or phase-conjugation reflectivity, and are important also in the study of phase conjugation and self-pumped phase conjugation involving thin nonlinear media where diffracted beams are generated.

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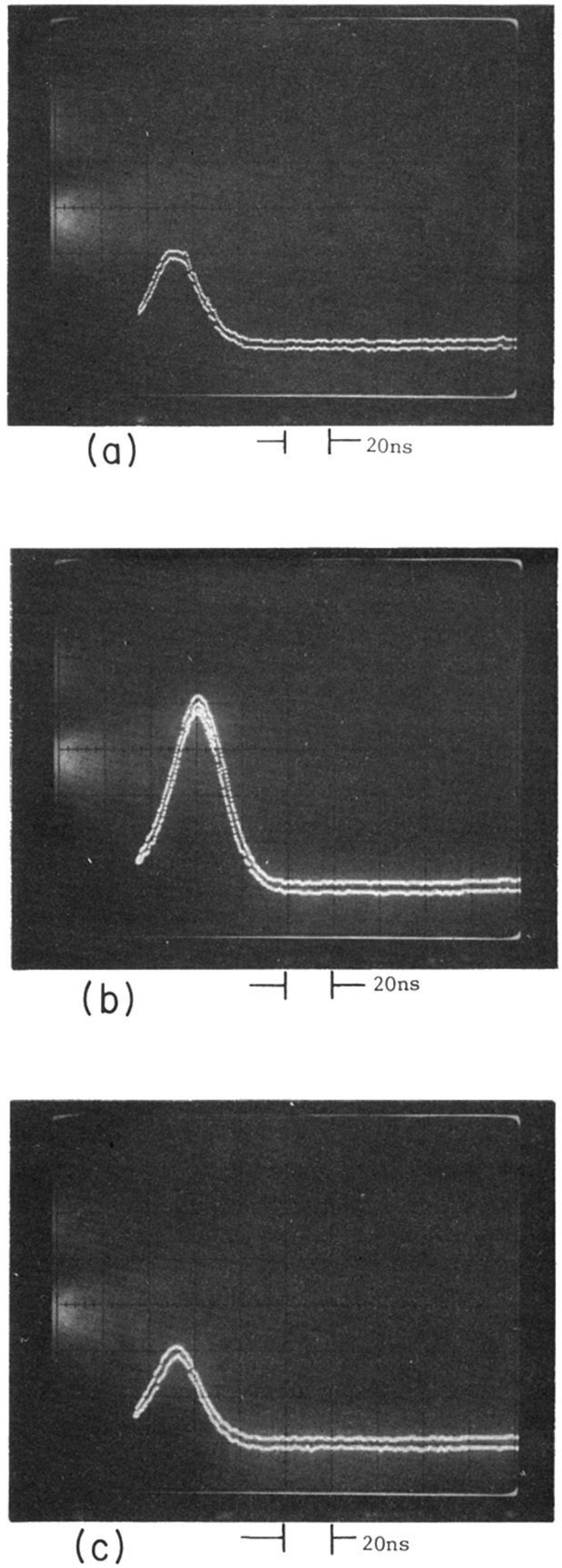

FIG. 6. (a) Oscilloscope trace of the probe pulse alone. The double trace is an artifact of the transient used in the pulse detection. (b) Oscilloscope trace of the probe beam in the presence of the forward-propagating pump beam (I_f) alone, showing an amplification of the probe beam. (c) Oscilloscope trace of the probe beam with both pump beams on (I_f and I_b), showing diminished gain.